The work presented in this thesis has two parts. The first part deals with methodological developments in the area of solid-state NMR, relevant to the study of partially ordered systems. Liquid crystals are best examples of such partially ordered systems and they are easily oriented by the magnetic field used for the NMR study. They provide spectra rich in information useful for the study of structure and dynamic s of the oriented molecule. Dipolar couplings and anisotropic chemical shifts are relatively easy to obtain for these systems. However, the methodologies used for extracting the required information are constantly undergoing change, with newer ideas being used for optimal use of the technique and increasing the sensitivity of the methodology. In this thesis, existing methods used for obtaining dipolar couplings from oriented liquid crystalline samples are examined in detail and conditions for optimal use of the methods are investigated. Different approaches for enhancing the sensitivity of the techniques are also proposed. Estimation of chemical shift anisotropy of carbons for a molecule that is used as a building block for several mesogens has been obtained and its utility for estimating the order parameters of the system have been examined. The second part of the thesis deals with the application of solid state NMR methods to the study of a number of novel liquid crystalline systems and for the estimation of dynamics, order and orientation of the mesogenic molecules in the magnetic field.

Chapter-2 deals with a detailed and systematic study to improve the sensitivity of cross-polarization based separated local field (SLF) NMR spectroscopy techniques such as PISEMA(Polarization Inversion Spin Exchange at the Magic Angle) and PDLF(Pro-ton Detected Local Field). The chapter has been further divided into three sections. Section-A describes the optimization procedure for cross-polarization period for reducing zero-frequency peaks in SLF experiments. Polarization Inversion(PI) is one of the important components of PISEMA and plays a crucial role in enhancing the dipolar cross-peaks and suppressing the axial-peaks. Shortening this period has the advantage of less r.f. power input into the system, thus less susceptibility to sample heating. Therefore it is crucial to arrive at the optimum condition for which maximum sensitivity and resolution are obtained. A detailed experimental investigation of the role of the initial po1arization period has been carried out for two different samples of static oriented liquid crystalline material at two different temperatures and a contact time of 2ms has been found to be optimal for such samples. Insection-B of this chapter, the initial preparation period of the experiment is considered as a possible means of increasing the sensitivity of the experiment. Thus the use of cross-polarization via the dipolar bath by the use of a diabatic demagnetization in the rotating frame(ADRF-CP) has been proposed to be incorporated into PISEMA. To understand the CP dynamics, magnetization in double-and zero-quantum reservoirs of an ensemble of spin-1/2 nuclei and their role in determining the sensitivity the experiments have been theoretically examined. Experimentally, a modification incorporating ADRF-CP is shown to result in enhancement of signal-to-noise by as much as 90% in the case of rigid single crystals of a model peptide and up to 50% in non-rigid, partially ordered liquid crystalline systems. In section-C another useful SLF technique known as PDLF spectroscopy has also been examined. In this case a sweep of one of the r.f. amplitudes(RAMP-CP),rather than

ADRF is found to work well. The reason for the different behaviours has been discussed.

Chapter-3 highlights two experimental approaches used to extract the chemical shift anisotropy(CSA) tensor information from rotating solids. Section-A is devoted to the measurement of the CSA values of thiophene by using MAS side band analysis, by extracting the principal values from the intensities of just a few spinning side bands. Experiments have been performed on thiophene-2 carboxylic acid and thiophene-3 carboxylic acid samples and the carbon CSA values have been obtained. In section-B, CSA values of carbons of the core unit of the liquid crystal4-hexyloxybenzoic acid (HBA) have been obtained by using the recoupling pulse sequence SUPER(Separation of Undistorted Powder patterns by Effortless Recoupling).HBA belongs to an important class of thermotropic liquid crystals which are structurally simple and often used as starting materials for many novel mesogens. As this molecule could serve as an ideal model compound, high resolution ^{13}C NMR studies of HBA in solution, solid and liquid crystalline phases have been also undertaken. The CSA values obtained from the 2D SUPER experiment showed good agreement to those computed by DFT calculations. The CSA values were used for obtaining the order parameter of the system at different temperatures. These matched well the order parameter obtained from the ^{13}C-^{1}H dipolar couplings in the nematic phase determined by SLF spectroscopy at various temperatures. A knowledge of the CSA of the carbons is thus very useful, as they can be used for gaining knowledge about the system from the chemical shifts obtained from a simple 1D spectrum.

In chapter-4, 1-and 2-dimensional ^{13}CNMR techniques have been utilized to obtain extensive information about some novel mesogenic molecules. Four molecules of different structure and topology have been taken up for study. These molecules have the following features. Mesogen-1 has a terminal hydroxyl group. Such systems with further modification can result in mesogenic monomers for side chain liquid crystalline polymers. Mesogen-2 has a dimethyl amino group at one end and has three phenyl groups connected by appropriate linking units that form the core. In the third case, mesogen-3, the terminal hydroxyl group of mesogen-1 is replaced with a hydrogen such that ^{13}C-^{1}H dipolar couplings provide directly information on molecular ordering and orientation. In the fourth case, mesogen-4, the core is built with four phenyl rings. Here the fourth ring is linked to other three rings via a flexible chain unit. In each of these cases the 2DSLFNMR experiments have been carried out where ^{13}C-^{1}H dipolar couplings as well as ^{13}C chemical shifts were used for obtaining the order parameters of various segments of the molecule. The data provide useful insight into the phase behaviour, ordering and orientation of the molecules.

Chapter-5 discusses the applications of the natural abundance ^{13}CNMR techniques to thiophene based mesogens, that have the potential for use in molecular electronics material. Typically, these molecules consist of phenyl rings appropriately connected by linking units with thiophene. Different core units as well as different linking units to thiophene have been considered. The six mesogens

thus obtained have been investigated in detail using 1D and 2D NMR methods. ^{13}C-^{1}H dipolar couplings have been used to obtain ordering information, that show interesting correlation to the molecular orientation and dynamics.